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Luminescence and upconversion process in Er^{3+}:TeO_2-Na_2CO_3 glasses

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Abstract : The photoluminescence properties of Er^{3+}-doped alkali telluride glasses are investigated. The matrix system is $80Te_2O_2$ -$20Na_2CO_3$-Er_2O_3 .We grew glasses at varying concentrations of Er from 0.5% to 2.5% The Stokes luminescence characteristics under 380nm excitation have been studied and intense green emission was observed from samples of Er^{3+} doped glassy material Infrared to visible upconversion has been observed under IR at 800nm excitation. Visible upconversion center at 540nm corresponding to the $^4S_{3/2} \rightarrow {}^4I_{15/2}$ in Er . sharp intense green emission occurs when concentration of Er_2O_3 is about 1 5 mol% Excited state absorption is discussed as possible mechanisms for the upconversion process under IR excitation

Keywords : Upconversion , erbium, tellurium oxide

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Erbium ion has been recognized as one of the most efficient rare-earth ions for obtaining laser emission, frequency upconversion , as well as to be used in optical amplifiers, when doping in different hosts. Vitreous hosts are desired to possess a minimum absorption coefficient within the wavelength range of interest and capability of incorporating high rare-earth concentrations. Frequency upconversion has been extensively investigated for erbium- doped host materials such as fluoride, chloride, germanosilicates, lead silicate, fluorozirconate and aluminosilicate glasses [1-4]. In contrast to the more extensively investigated oxide and halide compound, a higher number of excited states are available for luminescence process and upconversion process. This is because multiphonon relaxation is strongly suppressed in this heavy halide system due to their low phonon energies. However, they have a major drawback; they are all very sensitive to moisture and therefore, need of handling in dry boxes and protection during measurement and storing.

The Te_2O_2 -Na_2CO_3-host is a suitable glass host for all-solid state upconversion luminescence, as it is composed of relatively heavy atoms and provides a low multiphonon loss compared to other oxide glasses [5-8]. In this work, we have prepared Er-doped alkali telluride glasses and studied their emission spectrum of Stokes and upconversion luminescence.

The samples were prepared by mixing together glass forming TeO_2 (Aldrich99%)with Na_2CO_3 and higher purity Er_2O_3 (Fluka99.9%) powders as starting materials. About 5g batch of materials which were dried by heating at appropriate temperatures, were melted in alumina crucible at 800-850^{0}C about 30-45 min in muffle furnaces. The sample was then obtained by pouring quickly the melt onto a metal sheet. After annealing at 200^{0}C for 10min, samples were cooled to room temperature. The sample used in this work were of the general composition $80TeO_2$ + $(20–x)$ Na_2CO_3 + (x) Er_2O_3 where $x = 0.5,1,1.5,2,2.5$ mol%.

The fluorescence was measured with a Hitachi-650-10S Fluorescence Spectrophotometer . An Xe lamp (150W) was used as the UV (at 380nm) and IR (at 800nm) excitation source .

Figure1 shows the fluorescence spectra of Er^{3+} at 1.0 and 1.5mol% -doped TeO_2 : Na_2CO_3 system under 380nm excitation. The fluorescence profile largely change when excitation level $^4G_{11/2}$ is pumped (highest peak at 1.5 mol% of Er). In particular, the spectral profile in green region (520 -560 nm) consists of not only $^2H_{11/2} + {}^4S_{3/2} \rightarrow {}^4I_{15/2}$ transition but also $^2H_{9/2} \rightarrow {}^4I_{13/2}$ transition as shown in Figure 2 (a) state $^4G_{11/2}$or $^2H_{9/2}$ is

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pumped. The strength of emission intensity of the system is decided by the following two factors: one is the multiphonon relaxation rate and another is the line strength of each transition. The former are only dependent on phonon energy of the host materials. On the other hand, the variation of the latter with the host composition is generally small.

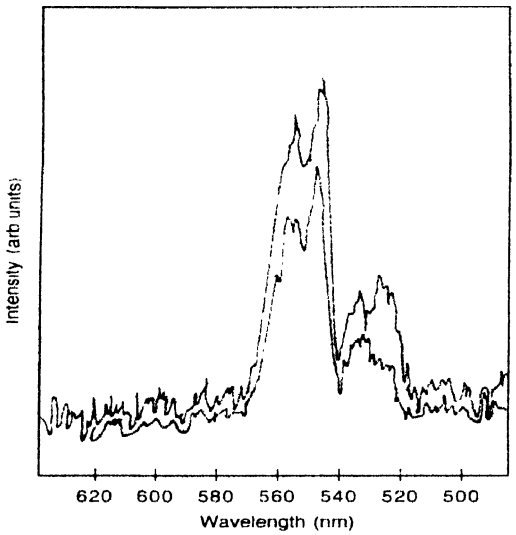

Figure 1. Fluorescence spectra of Er^{3+} in TeO_2 Na_2CO_3 glasses under ($^4G_{11/2} \rightarrow {}^4I_{15/2}$) 380nm excitation

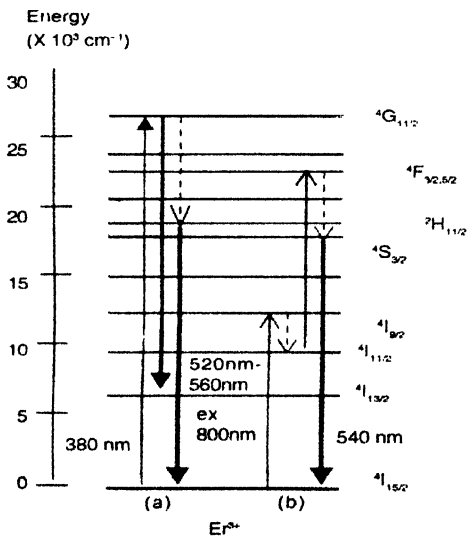

Figure 2. Energy level diagrams of Er^{3+} ions-doped glass for (a) normal fluorescence transition process and (b) upconversion process.

The upconversion luminescence of Er^{3+} doped tellurium glasses has been investigated with varied Er^{3+} ions concentration from 0.5mol% to 2.5 mol%. It has been observed (Figure 2(b)) that the same green emission bands at 540nm, although the intensity of the green emission varied with Er^{3+} concentration. Figure 3 shows the green emission at 1.0 and 1.5 mol% of Er^{3+} doped 80 TeO_2 : 20Na_2CO_3 glassy material under 800nm excitation. The strong emission bands around 530-550nm can be assigned to the $^4S_{3/2} \rightarrow {}^4I_{15/2}$ transition due to back energy - transfer from Er^{3+} ions at 1.5 mol% .

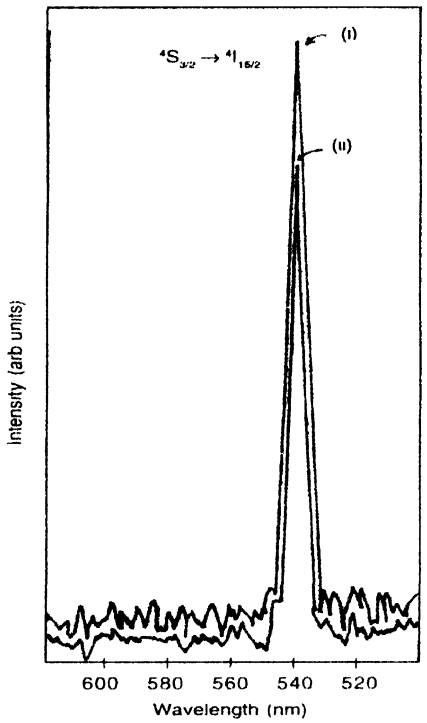

Figure 3. Upconversion luminescence spectra of Er^{3+} ions (i) 1.5mol% and (ii) 1.0mol%- doped glassy samples under 800nm excitation

Under 800nm pumping, Er^{3+} ion is excited initially from $^4I_{15/2}$ to $^4I_{9/2}$ state through the ground state absorption (GSA) process, and they decay to $^4I_{11/2}$ state due to multiphonon relaxation process. The same ion absorbs further a second photon and is promoted to $^4F_{3/2}$ and $^4F_{5/2}$ states under excited state absorption (ESA) [8,9]. Finally, the ion decays to $^2H_{11/2}$ state, and $^2H_{11/2}$ state can also decay to $^4S_{3/2}$ state due to multiphonon relaxation process, and $^4S_{3/2} \rightarrow {}^4I_{15/2}$ transition giving green emission at 540 nm, as shown in Figure 2 (b). Narrow line width is generally preferable to glass laser because the stimulated emission cross-section is inversely proportional to the effective line width of the transition.

In conclusion, we have experimentally observed luminescence and frequency upconversion fluorescence in Er^{3+} ions doped $30Te_2O_2 - 20Na_2CO_3$-Er_2O_3 glass. In the emission spectrum of both processes, intense visible green fluorescence centered around 520nm, 545nm and 540nm under 380nm and 800nm excitation respectively are observed. At high concentration *i.e.* more than 1.5 mol % of Er^{3+}, both the upconversion luminescence and Stokes fluorescence intensity

decrease. This upconversion requires two-photon absorption (ESA) followed by multiphonon relaxation process of the dopant Er^{3+} ions. The most intense green emission occurs when concentration of Er_2O_3 is about 1.5 mol%.

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